MICROWAVE ASSISTED REACTIVE BRAZING OF CERAMIC MATERIALS

The present application claim the benefit of the priority filing date of provisional

5   patent application No. 60/455,204 filed on 07 March, 2003, hereby incorporated, in its

entirety, by reference.

FIELD OF THE INVENTION

This invention relates in general to the field of ceramics and in particular to the

10   field of bonding high strength or high temperature ceramics.

BACKGROUND OF THE INVENTION

With the advent of new high tech materials, such as high temperature/high

strength ceramics, the efficient joining of these materials is a major technical issue and

15   very important to wide use of these materials. Many of these materials cannot be

produced in large component sizes. Consequently, use of such materials in large scale

devices requires joining of smaller components into assemblies of the required size.

Also it is difficult, if not impossible, to make high performance ceramic materials

in complex shapes. The joining of simpler shapes on the other hand allows the

20   preparation of more complicated shaped high performance ceramic parts, provided that

the joints and surrounding regions exhibit, at all required operating conditions, the same

strength and durability as the basic ceramic material.

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Another difficulty with large scale ceramic assemblies or components can be their relative fragility compared to other metal or polymeric structures. Shipping of such large assemblies from the site of manufacture to the site of use may well result in damage to the assembly and large replacement cost. One remedy for these problems is a robust

5   capability for joining high temperature ceramics. This capability should include techniques for joining subassemblies in a production facility, but should also be capable of being used in a field setting, at the site of use.

One example of such a requirement is the production of large silicon carbide (SiC) heat exchanger assemblies for hot gas recuperation. This requires assembly of

10   various tube segments, couplers, manifolds and other SiC and metal components into the large heat exchanger assembly. The joints must have sufficient strength to withstand the process pressures and handling during installation, and must also be gas tight. Another example is an accelerator application where there is a need to assemble a series of hot - pressed, high purity alumina tube segments, each about 3-4 cm in length, into a meter

15   long dielectric-loaded accelerator assembly. In this case the joints need to have similar dielectric properties as the alumina, including very low loss and high breakdown voltage, and a pore free structure.

Another application is for joining ceramics plates to create complex structures such as conformal ceramic armor. If a low cost and efficient way to join ceramic plates

20   such that the joint area shares similar physical and chemical characteristics with the base materials was available, complex, light weight multi-component assemblies could be assembled to conform to structures such as seats, engines, gearboxes or etc.

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Hence, there exists a need to be able to join high performance ceramics, where the

joint area shares the same or similar physical and chemical characteristics with the base

materials.

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SUMMARY OF THE INVENTION

A multi-component assembly featuring a structure with joints that share physical,

chemical and electrical characteristics with the base materials and a process for creating a

multi component structure by bonding base material parts using localized microwave

10   energy and featuring the steps of:

Applying a thin coating of a joining material to each surface of the base materials

to be joined; disposing each of the base materials such that the coated surfaces of the

base materials being joined are in intimate contact in the desired alignment, and the

pressure necessary to maintain the intimate contact and the desired alignment when

15   joining the base materials and creating the desired component assembly; heating the

joint area with a microwave beam applied to the surfaces of the base material being

joined and focusing or diffusing the microwave beam to achieve localized heating of the

joint area; heating the joint area to an initial joining temperature, wherein the joining

material softens and fills physical discontinuities between the surfaces of the base

20   materials being joined; rapidly heating the joint area to the reactive temperature of the

joining material and the base materials; maintaining the joint area at the reactive

temperature for a short interval to allow for the interdiffusion of the base and joining

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materials and formation of a homogenous joint region; rapidly cooling the joint area to a

recrystalization temperature and maintaining the joint area at the recrystalization

temperature for a predetermined period; slowly cooling the joint area to room

temperature, wherein the resulting component structure features joint region having

5   similar physical, thermal and electrical characteristics as the base materials.

BRIEF DESCRIPTION OF THE DRAWINGS

FIGURE 1 shows the joint structure featuring the joining material applied to the

surface of the base materials being joined.

10   FIGURE 2 shows the joint structure featuring the joining material and the base

materials being joined during the initial heating phase of the joint area via a localized

microwave beam.

FIGURE 3 shows the joint structure featuring the joining material and the base

materials being joined during the reactive temperature phase.

15   FIGURE 4 shows the finished joint structure after recrystalization.

FIGURE 5 shows an example configuration of an apparatus for performing the

microwave seamless joint bonding process at the heating stage.

FIGURE 6 shows a heat treatment schedule in accordance with an exemplary

method of the invention.

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DETAILED DESCRIPTION

Prior to a detailed description of the illustrations a brief overview of the process

and of the resulting structure may be appropriate: Disclosed is a method for joining

material to efficiently create multi component assemblies in which the joints between the

various components have virtually the same physical, chemical and electrical

5   characteristics as the base materials. The process for creating such multi component

structures uses a thin layer of a selected joining material applied to the juncture and

localized microwave energy focused thereon to bond the component materials, and to

diffuse away the joining material into the base material.

A thin coating of the joining material is applied to the surfaces of the base

10.   materials being joined. The base materials are disposed in relation to each other such

that the two surfaces of the base materials being joined are in contact with each other and

feature the desired alignment, and pressure necessary for maintaining the contact and the

alignment required for the desired component assembly. The actual magnitude of the

pressure applied during joining makes a minor contribution to the amount of materials

15   diffusion that causes the joining during the heating process. Temperature and duration of

the heating and cooling cycles are its primary determinants.

A microwave beam is focused on the joint area. The joint area is the area defined

by the surfaces of the base material being joined. The microwave beam incident on the

joint area is focused to achieve localized heating of the joint area. The surfaces beyond

20   the joint area remain cool.

The joint area is heated to an initial joining temperature, wherein the joining

material softens and fills physical discontinuities between the surfaces of the base

materials being joined. The materials can be held at this temperature for a short interval.

The joint area is then rapidly heated to the reactive temperature of the joining material

and the base materials. The joint area is maintained at the reactive temperature for a

short interval to allow for the reaction and interdiffusion of the base and joining materials

5   and for the subsequent formation of a homogenous joint region.

Next the joint area is rapidly cooled to a recrystalization temperature, typically

well below the initial softening temperature, and the joint area is maintained at the

recystalization temperature for a predetermined period. This allows the joint region,

now featuring the base materials regions interdiffused with each other and with the

10   joining material, to form a homogeneous stable physical and thermal structure having

uniform grain sizes. This process can be used to create very complex structures through

sequential joining since the previous made joints are not significantly heated. Even if

their temperature is increased, their properties would not be affected any differently than

those of the base material, since the joining material by that time is expected to have

15   diffused away.

With reference to the figures in which like reference numbers denote like

elements, Figure 1 illustrates an example joint structure featuring the joining material

applied to the surface of the base materials being joined. The process for bonding the

base materials and creating the seamless joint structure employs localized microwave

20   energy for heating. The microwave energy is focused on the joint area, causing the

localized heating of the selected area only. The other surfaces of the base materials

remain near the ambient temperature.

Figure 1 shows an example embodiment of the joint structure during the initial

steps of the joining process. Base materials, 150, 170 are disposed to be in intimate

contact with each other after a joining material 140 is applied to the joint area 160 of the

base materials 150, 170 being joined.

5   Generally the base materials 150, 170 comprise high temperature ceramics which

may be similar or dissimilar in chemical composition, shape and attributes. In a

preferred embodiment these base materials are high temperature ceramics having a purity

of greater than 99%. The base materials may be high temperature ceramics in the form

of high purity oxide materials, however oxides are not required. In addition, the base

10   material may be dissimilar materials.

As illustrated in Figure 1, a thin coating of a joining material 140 is applied to

each surface of the base materials, 150 and 170 being joined. The coating is applied to

the joint region 160, specifically to the joint surfaces 130 of the base materials to be

joined.

15   The joining material is a frit or reactive braze selected such that at a

predetermined temperature the constituents of the joining material will chemically react

and interdiffuse with the base material. As a result of the frit or braze constituents

diffusing away into the base materials, their concentration is reduced to undetectable

levels, so that only the high temperature ceramic material remains, including in the joint

20   area. The joining material is chosen to be chemically reactive with the base material at a

temperature below that of the thermal degradation threshold of the base material.

In an example embodiment, wherein the base materials are high temperature oxides, the joining material would preferably be an oxide glass frit. The use of such joint material permits assembly with minimal bonding pressure, since the glass initially softens and flows during the bonding process. The use of an oxide glass frit also relaxes

5   the requirements for fit and finish in joints - surface flatness and roughness, since the reactive glass frit can fill surface irregularities in the joint. With the use of localized heating, the process can be used to create complex, multi surface structures through sequential heating, since previously made joints are homogenized by the diffusion process to have essentially the same composition and high temperature tolerance as the

10   original base ceramic.

Referring again to Figure 1 and to Figure 5, each base material 150, 170 is disposed such that the surface of the base materials being joined 130 are in contact, with the desired alignment, and pressure necessary for joining the base materials and creating the desired component assembly. Figure 5 illustrates an example system as employed to

15   create a multi component assembly according to the method described herein. Fixtures 580, 580' are employed to dispose each piece of base material 150, 170 such that the surface of the base materials being joined are in contact with the desired alignment, and pressure necessary for joining the base materials 150, 170 and creating the desired component assembly.

20   The fixtures 580, 580' may be low temperature-type fixtures and are used to position the respective pieces of the base materials 150, 170 with the desired alignment and pressure to create the desired joined structure.

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Once the base materials 150, 170 have been coated at the surfaces to be joined

and aligned, the next phase involves heating the joint area 160 with a microwave beam

220 applied to the surfaces of the base material being joined. The heating effect may be

controlled by focusing or diffusing the microwave beam 220 to achieve localized heating

5   of the joint area 160.

Figure 2 illustrates the joint structure featuring the thin layer of the joining

material 140 and the base materials being joined 150, 170 during the initial heating phase

of the joint area 160 via a localized microwave beam 220.

Microwave beam heating of materials offer a number of advantages in material

10   processing and particularly in joining operations. One is much shorter processing times

than in conventional furnaces. With focused millimeter-wave heating of materials, only

the relatively small joint area is heated, minimizing effects of the thermal inertia of the

system. This permits very rapid temperature changes. Typically in microwave and

millimeter wave systems heating and cooling rates are only limited by two factors -

15   thermal shock of the workpiece and the ability of the workpiece to dissipate heat to the

environment during cooling.

Preferably, the beam is a millimeter-wave beam. In an embodiment, heating is

accomplished with a 83 GHz, gyrotron-based millimeter-wave processing system with a

superconducting magnet. The magnet is cooled by a cryogenic refrigerator to reduce

20   operating costs that can be associated with liquid helium cooling. The gyrotron is

capable of a maximum continuous output of 15 kW in a beam about 4 - 5 cm in diameter.

The beam exits the gyrotron through a BN window and is transmitted to the work

chamber via a metal transition, contaiing a secondary, sacrificial, TEFLON material

coated BN window. The window protects the primary gyrotron window, which should

maintain a high vacuum, from corrosive or reactive atmospheres or products of the

material processing. Within the work chamber, the beam can be manipulated using gold-

5   plated copper mirrors and reflective lenses, controlled remotely by a three-axis

manipulator outside the chamber via a vacuum and gas-tight feedthrough. The work

chamber can be constructed of non-magnetic stainless steel, has two access doors,

provision for exhausting gases from the chamber, and numerous feedthroughs.

Another major factor in the beam system employed is the ability to localize

10   heating. This is clearly impossible in a conventional furnace or in a microwave cavity

system. By employing optics as in the present method, one can focus the beam down to a

spot the size of a centimeter. The beam is focused, defocused or shaped by suitable

reflective optics. The optics may even be equipped with motorized drives, if desired.

This localization of the microwave beam has two important consequences. The first is

15   that the heating can be confined to the joint area in an assembly being joined by this

process, minimizing or eliminating heating of the remainder of an assembly. Confining

the heating to the joint area eliminates thermal damage to thermally sensitive sub-

elements of an assembly, while providing sufficiently high temperatures in the joint

region to achieve a high strength, refractory joint in high temperature base materials.

20   Another related effect which may be used to achieve even greater localization of

heating, is that with suitable dielectric properties, the incident millimeter wave beam can

be guided down the joint itself, with essentially no energy deposited adjacent to the joint.

This effect may be used to heat joints much smaller than the wavelength of the radiation used in current microwave generating "gyrotron" system.

Referring now to Figure 2 and with continued reference to Figure 5, localized microwave beam heating is applied to the joint area 160 of the base materials 150, 170.

5   The joint is heated to an initial joining temperature. As shown in figure 2, at the initial joining temperature the joining material 140 softens and fills physical discontinuities between the surfaces of the base materials 150, 170 being joined. In the example embodiment, the initial joining temperature is in the range of approximately 800 to 1200 degrees Centigrade depending on the materials selected. The initial joining temperature

10   is selected as a temperature at which the frit softens substantially. The temperature is held for a short interval, for example, for about 15 minutes for alumina material joined by a reactive oxide frit.

Next the joint area is rapidly heated to the reactive temperature of the joining material and the base materials. The reactive temperature is defined as the temperature at

15   which the joining material and the base materials chemically react. This temperature is generally greater than the initial joining temperature. The joint area is heated from the initial joining temperature to the reactive temperature at a rate of approximately 50 to 100 degrees Centigrade per minute. A suitable rate is about 100 degrees Centigrade per minute, or greater. In the example embodiment, the reactive temperature is

20   approximately 1400 to 1700 degrees Centigrade.

The joint is then maintained at the reactive temperature for a short interval to allow for the interdiffusion of the base and joining materials and formation of a

homogenous joint region. For example, the reactive temperature can be maintained for about 5 minutes.

Figure 3 shows an example joint structure featuring the joining material and the base materials being chemically joined during the reactive temperature phase.

5   As shown in the figure, base materials 150 and 170 are separated by a homogenous joint region 160 in which the constituents of the base materials and the joining material become interdiffused. Specifically base material 150 contains constituents represented by the letter "A" 151, while base material 170 contains constituents represented by the letter "C" 171. Base material 140 contains constituents represented by "B" 141. As

10   shown in the figure constituents 151 diffuse into the join area 160 and the base material 170 while constituents 171 from base area 170 diffuse into the joint area 160 and into the base material 150. The constituents of the joining material 141 simultaneously diffuse into the base materials 150, 170, facilitating the formation of a homogenous joint. In a preferred embodiment the joint area is maintained at the reactive temperature for an

15   interval of approximately 10 minutes or more.

Following the reactive temperature phase, the joint is allowed to cool rapidly to the recrystalization temperature. The recrystalization temperature is the temperature at which the joint region, which now has a homogenous composition, begins to attain a stable physical and thermal structure. The recrystalization temperature is well below the

20   intial softening temperature of the frit. The recrystalization temperature is maintained for a predetermined period or generally until an invariant crystal structure is formed in the

joint area. The recrystallization/annealing temperature is often 800 to 900 degrees

Centigrade.

The joint area is maintained at the recrystallization temperature for approximately

30 minutes, or until the joint area forms a stable physical and thermal structure. Once the

5   joint area forms stable physical and thermal structures, the new assembly is then allowed

to cool to room temperature. This process may be through atmospheric cooling or

through some type of forced cooling, the only requirement is that the assembly is not

cooled so quickly as to cause structural or chemical degradation of the base and joint

materials. The assembly can be cooled at about 50 degrees C per minute, for example.

10   The entire cycle time can be about 1.5 to 2 hours, in the example described herein.

Figure 4 shows the joint substructure after the recrystalization/annealing process.

As one can see from the figure the joint area 160 features a seamless external surface,

sharing a near identical chemical and physical structure with the base materials 150, 170.

Once the joined structure or component assembly is created, the process may be repeated

15   as many times as necessary allowing subsequent component assemblies or base materials

to be added to create the desired multi-component structure.

After a completed joining operation, constituents "B" 141 are fully diffused into

base materials 150 and 160. Their concentration in the joint region 160 is diluted to the

extent that they have only a negligible and undiscernible effect on the properties of the

20   joint. The joint then contains only homogenized constituents of the high performance

base materials "A" 151 and "C" 171.

An additional benefit of use of localized heating by a directed microwave beam during the heating processes is that metal fixtures used to position and hold each base material do not generally reach a temperature above 100 degrees Centigrade. Generally, when other types of heating sources are used, heating effects cannot be contained to only

5   the joint area, thus special heat resistant fixtures must be employed such as fixtures formed from ceramics or some type of heat resistant alloy. With localized microwave beam heating, low cost, low temperature fixtures can be used. Once the joined structure or component assembly is created, the process may be repeated as many times as necessary allowing subsequent component assemblies or base materials to be added to

10   create the desired multi-component structure.

An advantage of using reactive, oxide glass frits is it permits assembly with minimum bonding pressure, since the glass initially softens and flows during the bonding process. Another advantage is that it relaxes requirements for fit and finish in joints - surface flatness and roughness, since the reactive glass frit can fill surface irregularities

15   in a joint. Further, with the use of localized heating, these processes can be used to construct complex assemblies through sequential joining, since previously made joints are not significantly heater. High strength joints can be produced when the glass frit chemistry is properly tailored to the materials being joined. In addition, the resulting joint will have high temperature properties similar to those of the ceramics being joined.

20   This characteristic is useful for systems that require the joined ceramic product to be used at elevated temperatures.

Moreover, the methods outlined herein, when joining oxide ceramics, can be conducted in ambient air and no special processing atmosphers are required. For non-oxide materials, such as SiC, for example, processing can be accomplished in an inert atmosphere such as argon or nitrogen.

5   Further, the methods disclosed herein also allow some limited stress relief during process, e.g., from joining of dissimilar materials, occurring in the presence of viscous glassy phases in the joint during the processing.

Localization of heating in a beam system has advantages in terms of fixturing and instrumentation for processing and joining. Because the effect of the beam is confined to

10   a small region on the workpiece, typically 2 - 5 cm in diameter, and the system can contain a water load to absorb any radiation not used to heat the workpiece, there is considerable freedom in the location and types of other devices in the work chamber. For example, the system can also include unshielded fine gage thermocouples (typically with glass fiber or TEFLON material insulation) for temperature measurement.

15   For purposes of example two or more thin pieces of high purity Alumina, such as WESGO 995, may be joined using the above method to form a complex compound structure. This alumina is of a very high purity, greater than 99.5%. The surfaces of the pieces of alumina to be joined are held together in intimate contact at a low pressure using low temperature fixtures such as those illustrated in Figure figure 5. The joint

20   surfaces are thinly coated with a glass frit. In this example embodiment, the frit material selected is a Calcia/Boria/Silica compound, such as the Fusion 588 glass frit. The frit comprises $CaO \cdot B_2O_3 \cdot SiO_2$. This material was chosen because of it becomes chemically

reactive with the base alumina at a temperature as low as 900 degrees C and the frits

softens at even lower temperatures.   Further, all the constituents are reactive with

aluminum and have refractory products: aluminum borate, mullite, and calcium

aluminate. The frit compositions melt at temperatures ranging from about 1000 degrees

5   C to about 1400 degrees C.

A localized millimeter wave beam is focused on the joint area, and the joint area

is initially heated to approximately 1200 degrees Centigrade. This is the initial joining

temperature. At the initial joining temperature the frit material softens and fills physical

discontinuities between the surfaces of the base alumina being joined.

10   Next the joint area is rapidly heated to the reactive temperature, which for

purposes of this example, is approximately 1700 degrees Centigrade. The joint area is

heated from the initial joining temperature to the reactive temperature at a rate of

approximately 100 degrees Centigrade a minute. Once the joint area reaches the reactive

temperature, the joint area's temperature is maintained at a constant 1700 degrees

15   Centigrade for approximately 2 minutes. This provides ample time for the alumina and

frit materials to interdiffuse.

The joint area is then cooled to the annealing/recrystalization temperature. For a

structure using the selected alumina and a $CaO \cdot B_2O_3 \cdot SiO_2$ frit material this temperature is

20   approximately 900 degrees C. The joint area is maintained at this temperature for

approximately 30 minutes.

The joint area is then allowed to cool to room temperature. The two pieces of

Alumina are now joined creating an Alumina compound structure featuring a

homogenous joint which shares physical and chemical characteristics very similar to the

base alumina.

5   Once the alumina compound structure has been allowed to cool, complex multi-

component assemblies may formed by joining the alumina compound structure with

another base material or compound substructure using the above method.

Commercial frit compositions are available from several sources that are suitable

for joining, with some modification of their heat treatment schedules. In some cases,

10   custom made formulations and heat treatment schedules can be developed, based on the

phase diagrams of the materials involved.

In one embodiment, the materials to be joined are lead-zirconite-titanate (PZT)

materials. In this embodiment, the joining material is a submicron TiO_2 material filled

with silver particles. PZT segments can be joined with a TiO_2/Ag reactive braze

15   according the above-described method using an 83 GHz gyrotron generated beam. The

method produces a strong bond between the segments, using temperatures high enough to

melt the silver while affecting very little of the adjacent material. For the PZT segments,

it is desired to produce high strength, fatigue resistant joints between poled PZT

segments, where the bulk of the segments would not be taken above 300 degrees C, in

20   order to avoid depoling the PZT segments. A PZT joint formed wtih the 83 GHz beam is

about 10 - 15 µm in width, with a heat affected zone of only 100 µm in width.

Oxide frits can be purchased or developed for other oxide ceramics, and there are

analogous formulations available for non-oxide ceramics such as nitrides. In the case of

carbides, particularly silicon carbide, similar techniques are possible using reactive

brazes containing Si and precursors for SiC and C.

5   Figure 8 shows a reactive oxide glass braze joint in alumina with heat treatment

to recrystallize the joint region, accordance with an embodiment of the invention.

Figure 9 shows a optical micrograph illustration of a joint in high purity alumina with

heat treatment to recrystallize the joint region, in accordance with an embodiment of the

invention. Figure 10 shows a scanning electron micrograph illustration of a joint region

10   in high purity alumina with heat treatment to recrystallize the joint region, in accordance

with an embodiment of the invention.

In an exemplary embodiment, the method and system for bonding the materials is

based on an 83 GHz gyrotron with superconducting magnet. The magnet is cooled by a

cyrogenic refrigerator to reduce the high operating costs associated with liquid helium

15   cooling. The gyrotron is capable of a maximum continuous output of 15 kW in a beam

about 4-5 cm in diameter. The beam exits the gyrotron through a BN window and is

transmitted to the work chamber via a metal transition, containing a secondary,

sacrificial, TEFLON® coated BN window, which protects the primary gyrotron window,

which must maintain a high vacuum, from corrosive or reactive atmospheres or products

20   from material processing. Within the work chamber, the beam is manipulated using gold-

plated copper mirrors and reflective lenses. These can be controlled remotely by a three-

axis manipulator outside the chamber, with vacuum and gas-tight feedthrough. The work

chamber is constructed of non-magnetic stainless steel, has two access doors provision

for exhausting gases from the chamber, numerous feedthroughs. The chamber is both

vacuum-tight and will withstand overpressures of about 1 atm. The system has been

operated with a range of processing atmospheres - vacuum, air, nitrogen, argon, and

5   nitrogen-hydrogen mixtures. A non-outgassing water load is placed in the chamber to

absorb any millimeter-wave radiation not absorbed in the workpiece. Process

instrumentation that is placed in the work chamber and read out through feedthroughs

includes three thermocouples of Type K or S, and a low temperature (400°C) infrared

thermometer. A two color optical pyrometer is used to measure temperature optically at

10   the workpiece, sighting through a shielded viewport. The entire system is operated with a

PC-based LabView™ software system that incorporates numerous interlocks and safety

features to ensure safe operation of the gyrotron-based processing system. The

LabView™ system also provides for data acquisition and storage from the process

monitoring instrumentation and a record of the operating parameters of the gyrotron

15   system as well. With many types of processing, the automated operating system permits

conducting many types of processing in the system with a single operator.

Millimeter-wave beam heating of materials offer a number of advantages in

material processing and particularly in joining operations. One is much shorter

processing times than in conventional furnaces. With microwave or millimeter-wave

20   heating of materials, only the workpiece is heated, minimizing the thermal inertia of the

system. This permits very rapid temperature changes, provided that sufficient power is

available. Typically in microwave and millimeter wave systems heating and cooling rates

are limited by two factors - the thermal shock of the workpiece and the ability of the

workpiece to dissipate heat to the environment during cooling. Basically, heating can be

as rapid as the material being heated will tolerate. Heating rates as high as 100°C/min,

and up to 1000°C/min, are suitable. Cooling is controlled by the thermal mass and

5   conductivity of the workpiece, and its ability to dissipate heat to the work environment

by radiation, convection or conduction. Cooling rates for small samples can be of

100°C/min if the millimeter-wave power is simply turned off.

Another advantage of the gyrotron based millimeter wave beam system is the

ability to localize heating. This is difficult or impossible in a conventional furnace or in a

10   microwave cavity system. In contrast, the millimeter wave beam generated by the

gyrotron can be focused with optics down to a spot size of a centimeter. The beam can

readily be focused, defocused or shaped by suitable reflective optics. If the optics are

equipped with motorized drives, the beam can be rastered in a controlled manner across

a workpiece. This localization has two important consequences. First, the heating can be

15   confined to the joint region in an assembly being joined by this process, minimizing or

eliminating heating of the remainder of an assembly. This can eliminate thermal damage

to thermally sensitive subelements of an assembly, while providing sufficiently high

temperatures in the joint region (e.g., 1400-1500°C) to achieve a high strength, refractory

joint in high temperature materials. A related effect can be used to achieve even greater

20   localization of heating, in which, with suitable dielectric properties, the incident

millimeter-wave beam is guided down the joint itself, with essentially no energy

deposited adjacent to the joint. This effect can be used to heat joints much smaller than

the wavelength (3.6 mm) of the radiation used in the gyrotron system. In one example,

PZT segments are joined with a reactive braze (TiO_2/Ag), which produced a strong bond

between the segments, achieving temperatures high enough to melt the silver, while

affecting very little of the adjacent material. This provides the capability to produce high

5   strength, fatigue resistant joints between poled PZT segments, where the bulk of the

segments cannot taken above 300°C without depoling the segments. A joint was

produced between the PZT segments with a 10-15 µm wiidth and a heat-affected zone

(HAZ) of only 100 µm in width.

Localization of heating in a beam system also has large operational advantages in

10   terms of fixturing and instrumentation for processing or joining. Because the effect of the

beam is confined to a small region on the workpiece, typically 2-5 cm in diameter, and

the system can include a water load to absorb any radiation not used to heat the

workpiece, there is considerable freedom in the location and types of other devices in the

work chamber. For example, the system can include unshielded fine gage thermocouples

15   (typically with glass fiber or TEFLON® insulation) for temperature measurement. Only

EM shielding of the plastic thermocouple connectors is needed. This is conveniently

done with copper tape. A low temperature IR thermometer also can be employed in the

chamber, which only requires shielding of its lead with copper braid and a fine mesh

screen over the aperture to the optics.

20   An important byproduct of the beam heating is the freedom to use almost any

material for fixturing for providing alignment of pieces being joined and for applying

pressure to the joints. In a conventional furnace system, such fixturing would have to be

made either of superalloys or from high temperature ceramics, would be very costly to

produce, and usually would require a long lead time for acquisition. In a microwave

cavity system, this is virtually impossible because of the high electric fields present

throughout the cavity and the resultant arcing at conductive objects and heating of

5   insulating materials. In the millimeter wave energy gyrotron system described herein, the

fixtures can be steel and BN. BN is a convenient ceramic material that is relatively low

cost, easily machinable using conventional machining tools, and is non-reactive with

most other ceramics. Although steel fixtures can be custom built for application of

pressure, small machinists vises are also suitable.

10   A system for rotating components under the beam during millimeter-wave joining

processes is also possible, using mostly commercial components and a sealed DC

gearmotor with power coming into our chamber through a feedthrough for variable

rotational speed on the workpiece. Such a system in a conventional furnace system would

be extremely difficult and expensive to produce, and would require large costs to adapt to

15   different joint geometries.

In an experimental setup, two thermocouples measure the temperature of the vise

and the temperature at the lower surface of the alumina segments. The temperature at

the top surface is read via a two color optical pyrometer. Pressure is applied to the stack

of alumina segments through BN spacers and alumina tubular insulators. In addition,

20   fine tantalum foil is used as a radiation shield, both for electromagnetic and IR, to

confine the beam energy, effectively producing a mini-cavity around the workpiece.

This experimental arrangement permits heating the alumina joint segments to temperatures over 1500°C, with the metal fixturing never reaching more than 50°C.

Other advantages in joining high temperature ceramics derive from the use of reactive brazes to join the ceramics, for example, reactive oxide glass frits for joining

5   high temperature oxides. The use of such joint materials permits assembly with minimal bonding pressure, since the glass initially softens and flows during the bonding process. It also relaxes requirements for fit and finish in joints - surface flatness and roughness, since the reactive glass frit can fill surface irregularities in a joint. With the use of localized heating, the process can be used to construct complex assemblies through

10   sequential joining, since previously made joints are not significantly heated. High strength joints can be produced when the glass frit chemistry is properly tailored to the materials being joined.

In addition, with proper heat treatment, the joint materials can be recrystallized, leaving a refractory, crystalline joint, that should have high temperature properties

15   similar to those of the ceramics being joined. This is especially desirable since ceramics are frequently used in high temperature applications. When joining oxide ceramics, the process is conducted in ambient air; no special processing atmospheres are required. For non-oxide materials such as SiC, processing can be done in inert atmospheres such as argon or nitrogen. Finally, because of the presence of viscous glassy phases in the joint

20   during processing, there is the opportunity for some limited stress relief during the joining process, e.g., from joining of dissimilar materials.

Commercial frit compositions are available from several sources that are usable

for the purpose of joining, with some modifications of heat treatment schedules. In some

cases, custom made formulations and heat treatment schedules can be developed, based

on phase diagrams of the materials involved. In an exemplary PZT joint formed by a

5   method according to the invention, the braze was chosen based on several constraints.

One of these was reactivity with the PZT material and the ability to form intermediate

compounds. A second was the need to have a conductive joint in the final product. A

third was the requirement to have the particular dielectric properties in the braze material,

to produce a guided wave propagating down the joint region. These were all satisfied

10   with the use of a submicron TiO_2 material filled with fine silver particles. In the case of

alumina joints, a commercial oxide glass frit normally used for sealing of alumina

components can be selected, with a heat treatment schedule to provide for a strong,

refractory crystalline joint in the product. The $CaO-B_2O_3-SiO_2$ oxide frit selected is

reactive with alumina, contains constituents which diffuse into the alumina, while the

15   aluminum in the alumina diffuses into the frit, and has a relatively low softening point for

purposes of easy sealing. All of the constituents are reactive with alumina and have

refractory products: aluminum borate, mullite, and calcium aluminate. The frit contains

useful compositions that melt at temperatures ranging from about 1000 to 1400°C.

Similar oxide frits can be purchased or developed for other oxide ceramics, and

20   there are analogous formulations for non-oxide ceramics such as nitrides. In the case of

carbides, particularly silicon carbide, similar techniques are possible using reactive

brazes containing Si and precursors for SiC and C.

In joining various types of aluminum oxide materials with the millimeter-wave

beam system, suitable size specimens of the aluminas are sectioned from billets or other

stock and ground as necessary. The specimens can be used as saw cut, with no further

preparation of the mating surfaces. Alternatively, the joints can be made with the as-

5   fabricated surfaces on commercial aluminas. A frit suspension, typically either 20 or 40

v/o fit in butanol is prepared using a commercial fluorocarbon dispersant in small

quantities (<0.1%). The frit isdispersed in the butanol using small zirconia milling media

and mild agitation. The surfaces to be joined are then coated with this suspension and

immediately assembled into the final configuration. Assembly of specimens to be joined

10   can be done in small machinists vises, with clamping pressure adjusted to apply about 0.5

MPa pressure to the joint areas. The alumina joining specimens are thermally isolated

from the vises using alumina tubes, BN spacers placed between the alumina tubes and

joining specimens to inhibit bonding. The assemblies were allowed to air dry for about

one hour prior to processing in the millimeter wave system. A Type K or S thermocouple

15   for example, depending on expected process temperatures, is placed beneath the joining

specimens, with the thermocouple tip touching the bottom of the specimens. A two color

optical pyrometer is used to measure the upper surface temperature of the joint

specimens, at temperatures-above 700°C. Additional thermocouples can be used to

monitor the temperature of the fixturing and of the chamber environment. A metal foil

20   radiation shield cab placed around the specimens. A sufficiently large gap is left to

permit entry of the millimeter-wave beam and sighting of the specimen top surface by the

pyrometer.

In actual processing, the joining assembly and fixturing are placed in the

millimeter-wave work chamber and located relative to the position of the beam, with the

beam optics adjusted to provide the desired size and shape of the beam at the top surface

of the joining specimens. With this done, the specimens are subjected to selected heat

5   treatment schedules, as shown in Table 1 and Figure 6, for example.

In most cases, the specimens are brought up to an initial hold temperature at a rate

of 50-100°C/min. The initial hold temperature isselected as one where the glass frit

softened substantially. This temperature was held for about 15 minutes, followed by a

rapid ramp at 50-100°C/min. to an elevated temperature to permit interdiffusion and

10   interreaction between the fit and the alumina. After a short hold, ca. 5 minutes, at this

temperature, the temperature is rapidly reduced to a lower temperature, well below the

initial softening temperature, and held for a longer time, approx. 30 minutes, to permit

recrystallization of the products of the joining process. Following this hold, the assembly

is then cooled down at about 50°C per minute, or as fast as heat transfer allowed to room

15   temperature. The entire cycle time was approximately 1.5-2 hours. Following joining,

some assemblies are sectioned for optical and electron microscopy of the joint regions.

Initially joint strengths are estimated from the force necessary to separate joined

specimens using a wedge placed on the joint line.   Double lap shear strength specimens

can be typically fabricated in groups of three in one run to provide better statistics on

20   strength for specimens processed identically. These specimens can be tested to failure in

compression on a universal testing machine. The shear strength of the joints is calculated

from the maximum load obtained and the area of the joints.   The stress distributions in

the joints are such that the peak stresses are higher than the values calculated, and most

of the errors possible in this type of test will result in further lowering of the strength

values measured. In a series of experiments, various heat treatment schedules were

evaluated, together with various modifications of the fit suspension system. Optical

5   microscopy and scanning electron microscopy are used evaluate fracture surfaces, joint

morphology and microstructure, and joint chemistry is evaluated through EDAX.

As an example, joints can be produced in a technical grade alumina using a

simpler heat treatment schedule. These joints still contain significant amounts of residual

glassy phase in the joint region, but exhibit shear strengths of about 50 MPa (7 ksi).

10   This strength is comparable to what might be achieved using high strength structural

adhesives, but the reactive glass braze joints should be usable up to the softening

temperature of the residual glassy phases, approx. 1000°C. The residual glassy phase is

clearly evident in Fig. 8 below as a darker band approx. 50-100 μm in width. One feature

of note here is the effectiveness of the technique in filling the relatively rough surfaces on

15   the as-sawn surfaces used for joining.

With the proper heat treatment schedule, substantially higher joint strengths were

achieved, and the nature of the joints changed dramatically. Good strength results are

obtained in both technical grade aluminas and high purity (99.8%) aluminas. While the

joint region is somewhat darker than the bulk high purity alumina, due to the different

20   composition, the joint region is well crystallized. In many areas, the joint region is

indistinguishable from the adjacent alumina in the SEM.

In one series of experiments, joints are made to join low and high purity

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aluminas, where the impurity content of the low purity aluminas would make possible a better analysis of the interdiffusion occurring during joining. To facilitate microscopic and elemental analysis of the joint region, these samples are sectioned on about a 30° angle to provide about a 2:1 magnification of the joint region. Substantial interdiffusion

5   does occur during the joining process. This indiffusion is important to the success of the heat treatment in recrystallizing the joint subsequent to joining.

The strength results obtained in a number of these joining experiments are summarized in Table I below. This table shows the material, temperature profile, hold times and measured shear strengths. The three temperatures referenced as A, B and refer

10   to the initial hold temperature for braze softening and bonding, a second higher temperature for interdiffusion, and a third, lower temperature for recrystallization of the products, respectively.

Table I

	Material	# of tests	Temp	Time (min)	Shear Strength (MPa)
15	Coors AD995	4	B	15-18	23
	Wesgo AL 500 (94%)	3	A	15	38
		1	A-B-C	15/2/30	20
	Wesgo AL 300 (97.6%)	1	B	11.5	20
		3	B	15	59
20		2	B	35	42
	Wesgo AL 995 (99.5%)	3	B-C	2/30	20
		2	A-C	15/30	84

| Wesgo AL 995 (99.5%) | 2 | A-B-C | 15/4/30 | 102 |
| | 1 | A-B +200°C - C | 15/2/30 | 120 |

Of note above is that the best results, in terms of shear strength, are obtained with

5   the highest purity aluminas. These are typically the most difficult to join, because of their

refractory nature and their chemical inertness. It is also expected that, as mentioned

previously, these strengths will persist to very high temperatures, perhaps as high as

1400-1500°C, based on the nature of the compounds formed during the recrystallization

process. Also note that the strength values above, in the better cases, may be

10   underestimates of the true strength. Thus, the joints are probably as strong in shear as the

base material, with a nominal shear strength of about 200 MPa.

Although this invention has been described in relation to the exemplary

embodiments thereof, it is well understood by those skilled in the art that other variations

and modifications can be affected on the preferred embodiment without departing from

15   scope and spirit of the invention as set forth in the claims.